



## Raman determination of C concentration in silicate melt under pressure: carbon solubility in MORB and mantle melting scenario

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Understanding Earth degassing is fundamental in global studies of our planet history, as well as in studies of its recent climate. Degassing occurs mainly at Mean Ocean Ridges via exsolution of CO<sub>2</sub> vesicles in ascending tholeiite magma, and probably begins at some 30 km under the ridge. Therefore, a precise knowledge of how carbon solubility varies during ascent from the source region is mandatory, a process for which the effect of pressure remains poorly known. A pressure increase induces melt compression, known to diminish argon dissolution with respect to Henry's law at pressures above 10 kbar, but this effect is poorly documented for carbon where things are complicated by the transformation of CO<sub>2</sub> into carbonate ion, CO<sub>3</sub>(2-). Early experimental investigations on carbon solubility in various silicate melts up to 20-30 kbar have shown that Henry's law is not followed at high pressures.

We have performed an experimental study of C dissolution in basaltic melts. Samples were either a grinded natural MORB or a mixture of carbonate and oxide powders leading to an iron-free tholeiitic composition. In the first case, carbon was charged by adding calcium carbonate in excess to oversaturate the mixture in CO<sub>2</sub>; in the second case, carbonates were adjusted to give a total CO<sub>2</sub> concentration of 11 wt%. Samples were placed into a platinum capsule and submitted to pressures from 10 to 50 kbar, at temperatures of 1600°C-1700°C. Experiments were conducted using a belt - type piston press at LPMCN - Lyon, France. Most successful experiments show bubbles at the top of the capsule attesting the presence of a CO<sub>2</sub> gas phase, and hence an initial oversaturation in carbon. Analysis of dissolved C was performed using a micro-Raman apparatus at IPG Paris, France. Dissolved carbon appears as clear bands due to carbonate ions (an intense peak at 1100 cm<sup>-1</sup> and a doublet in the 1350-1600 cm<sup>-1</sup> region), molecular CO<sub>2</sub> being not detectable. Calibration of Raman spectroscopy for quantitative analysis was done by preparing standards at atmospheric pressure and analyzing them using a stable isotope mass spectrometer. The results show that carbon concentration increases steadily with increasing pressure, a behavior consistent with (rare) previous studies on basaltic melts. We also have performed molecular dynamics simulations to investigate the dissolution of CO<sub>2</sub> in a silicate melt. The calculated solubility is consistent with the data, which help understanding how pressure acts on fluid and melt, and yield insight into the details of how CO<sub>2</sub> and CO<sub>3</sub><sup>2-</sup> interact with the melt network. However, the fact that the carbon solubility in a MORB is continuously increasing with pressure is somewhat surprising, and will be discussed.

This work has shown that

- (i) Raman spectroscopy can be used to quantify C content in natural samples
- (ii) The C solubility measured in basaltic melt exhibits a behavior with pressure different from that exhibited by rare gases.
- (iii) Our results have important implications concerning the history of the atmosphere degassing and scenarios of mantle melting.